

REMARKS

Claims 1-4 and 6-11, as amended, remain herein. Claim 1 has been amended, and claim 5 has been canceled. Independent claim 1 has been amended to clarify that the moisture content of the micro balloons is less than 0.1% by mass, and to include the limitations from claim 5 in claim 1.

Applicants believe that the Amendment places this application fully in condition for allowance, and surely in better condition for any appeal. Accordingly, entry of this Amendment and allowance of all claims 1-4 and 6-11 are respectfully requested.

1. Claims 1-11 were rejected under 35 U.S.C. §102(a), or in the alternative, under 35 U.S.C. §103(a) over Hitachi Metals JP 2002-326881 (hereinafter JP '881).

The present U.S. application claims the benefit not only of the PCT international application, but also claims the benefit under the Paris Convention of the Japanese priority application filed March 29, 2002, which is several months earlier than the '02 publication date of the cited Japanese Patent Application Publication 2002-326881.

Submitted herewith, as attachment A, is a sworn English translation of applicants' Japanese Priority Application No. 2002-094113, which supports the claims rejected over the JP '881 publication. The English translation antedates JP '811 as a *de jure* prior art reference under U.S. law, thus mooted the rejection under §102(b) based upon JP '881.

Accordingly, applicants respectfully request withdrawal of this ground of rejection, and allowance of all claims 1-4 and 6-11.

2. Claims 1-8, 10 and 11 were rejected under 35 U.S.C. §102(b), or in the alternative, under 35 U.S.C. §103(a), over Inoue U.S. Patents 4, 871,693 and 4,698,317. Claim 5 has been canceled. Claims 2-4, 6-8, 10 and 11 depend from claim 1.

Applicants discovered a method of forming a cordierite porous body comprising providing cordierite forming materials comprising an Al source, an Si source, and an Mg source, and firing the materials to form cordierite, wherein the Al source and Si source are at least partially provided by inorganic micro balloons containing Al_2O_3 and SiO_2 , wherein a moisture content of the inorganic micro balloons is less than 0.1% by mass, and wherein the Si source and Al source comprise more than 90% by mass of the micro balloons and therefore an MgO content of less than 10% by mass.

Neither Inoue '693 nor Inoue '317 discloses that an Si source and an Al source comprise more than 90% by mass of micro balloons. Rather, Inoue teaches 10-65% MgO, and therefore, a total Si and Al content of 90% or less.

Thus, there is no disclosure in Inoue '693 or Inoue '317 that would have suggested applicants' claimed invention to anyone of ordinary skill in the art. Further, there is no disclosure or teaching in either Inoue '693 or Inoue '317 that would have suggested the desirability of combining any portions of those references effectively to anticipate or render obvious applicants' claimed invention. Accordingly, claims 1-4, 6-8, 10 and 11 are patentable over Inoue '693 and/or Inoue '317, and applicants respectfully request withdrawal of this ground of rejection, and allowance of all of those claims.

Serial No.: 10/509,155
Docket No.: 28953.7272

This application is now fully in condition for allowance and a notice to that effect is respectfully requested. The PTO is hereby authorized to charge/credit any fee deficiencies or overpayments to Deposit Account No. 19-4293 (Order No. 28953.7272). If further amendments would place this application in even better condition for issue, the Examiner is invited to call applicants' undersigned attorney at the number listed below.

Respectfully submitted,



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Date: October 30, 2006

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Attachment A



VERIFICATION

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do hereby verify that I am fully conversant with the Japanese and English languages and that attached translation signed by me is, to the best of my knowledge and belief, a true and correct English translation of the Japanese Patent Application No. 2002-094113.

DATED October 17, 2006

SIGNED 



PATENT OFFICE
JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the
following application as filed with this office.

Date of Application: March 29, 2002

Application Number: 2002-94113

Applicant(s): NGK Insulators, Ltd.

Commissioner,
Patent Office



[NAME OF DOCUMENT] APPLICATION FOR PATENT

[SERIAL NUMBER] WP04033

[FILING DATE] March 29, 2002

[ADDRESSEE] Kozo Oikawa

Commissioner of the Patent Office

[INTERNATIONAL PATENT CLASSIFICATION] B01D 39/20

B01J 35/10

F01N 3/02

C04B 35/00

[TITLE OF THE INVENTION] Method for Manufacturing
Cordierite Porous Body

[NUMBER OF CLAIMS] 5

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[Payment registered number] 009689

[Amount of payment] ¥21,000

[LIST OF SUBMITTED DOCUMENTS]

[Name of document] Specification : 1

[Name of document] Abstract : 1

[General power of attorney number] 9001231

[PROOF] Necessary

[Name of Document] SPECIFICATION

[Title of the Invention] Method for Manufacturing Cordierite porous body

[Scope of the Claim for Patent]

[Claim 1] A method of manufacturing a cordierite porous body using a cordierite forming material including an Al source, an Si source, and an Mg source and forming cordierite by firing,

characterized in that an inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part or all of the Al source and the Si source.

[Claim 2] The method of manufacturing the cordierite porous body according to claim 1, wherein a total content of the Si source and the Al source included in the inorganic micro balloon with respect to the whole inorganic micro balloon is 90% by mass or more, when the Si source is converted to SiO_2 , and the Al source is converted to Al_2O_3 .

[Claim 3] The method of manufacturing the cordierite porous body according to any one of claims 1 to 2, wherein a total content of a sodium compound and a potassium compound included in the inorganic micro balloon with respect to the whole inorganic micro balloon is 2% by mass or less, when the sodium compound is converted to Na_2O , and the potassium compound is converted to K_2O .

[Claim 4] The method of manufacturing the cordierite porous body according to any one of claims 1 to 3, wherein a melting point of the

inorganic micro balloon is 1400°C or more.

[Claim 5] The method of manufacturing the cordierite porous body according to any one of claims 1 to 4, wherein talc is used as a part or all of the Mg source.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to a method of manufacturing a cordierite porous body having characteristics which are preferable for a material constituting a filter, a catalyst carrier or the like mainly for purifying automobile exhaust gas.

[0002]

[Prior Art]

In recent years, a diesel particulate filter (DPF) which traps particulates discharged from a diesel engine has attract attention, and there has been a demand for a DPF capable of obtaining a high trapping efficiency with a low pressure loss. As the DPF, a honeycomb structure formed of cordierite has heretofore been used, and a porosity, pore distribution and the like of the honeycomb structure have heretofore been improved in order to obtain the above-described high trapping efficiency with the low pressure loss.

[0003]

In JP-A-9-77573 is described a honeycomb structure whose porosity and average pore diameter are increased and whose pore

distribution in a partition wall surface is stipulated, and in JP-A-11-333293 is described a honeycomb structure whose partition wall thickness is set to be not more than a predetermined thickness and whose porosity is increased.

[0004]

Moreover, in JP-B-7-38930 is described manufacturing of a high-porosity honeycomb structure by the use of particles which is as coarse as or coarser than predetermined as particles of both talc and silica components in a cordierite forming material, and in Patent No. 2726616 is described a honeycomb structure whose porosity is increased and whose pore distribution and surface roughness are stipulated. It is to be noted that in these conventional techniques, to raise the porosity, the cordierite forming material is formed into coarse and large particles, and graphite, wood powder, foaming agent or the like is added as a pore former.

[0005]

[Problems to be Solved by the Invention]

However, when the cordierite forming material is formed into the coarse and large particles, a cordierite forming reaction does not sufficiently proceed, and it has been difficult to achieve a low coefficient of thermal expansion. In a case where graphite is used as the pore former, permittivity of a formed body to which graphite has been added drops. When the added amount increases, it is difficult to uniformly perform drying by dielectric drying or

microwave drying. Further in a firing step, a firing time is lengthened at 800 to 1000°C, and therefore there has been a problem that rapid burning of graphite needs to be inhibited.

[0006]

Moreover, when starches and wood powders are used as the pore formers, a large amount of water needs to be added in order to form a clay into a predetermined hardness in a kneading step, and there has been a problem that an efficiency of a drying step is degraded. Further in a firing step, the starches and wood powders cause rapid burning and generate heat at 200 to 400°C, and it has been difficult to prevent firing cracks from being caused by this step. In this manner, in the conventional techniques, it has been remarkably difficult to increase a porosity to be not less than a predetermined porosity.

[0007]

Moreover, to obtain a higher porosity, for example, a porosity of 60% or more, a large amount of pore formers need to be also added. When a large amount of organic compound-based pore formers are added in this manner, amounts of an organic volatile substance, and a gas such as carbon dioxide generated in a degreasing (calcining) stage also increase, and a burning heat also increases. Therefore, defective portions such as cracks, tears and cuts, that is, defective places which do not exert any filter function and in which leakage of a fluid occurs are formed in an obtained calcined (degreased) body or fired body in some case.

[0008]

The present invention has been developed in consideration of these problems of the conventional techniques, and an object thereof is to provide a method of manufacturing a cordierite porous body having a high porosity, in which there is an extremely low possibility of generating defects causing leakage of a fluid, such as cuts.

[0009]

[Means to Solve the Problem]

That is, according to the present invention, there is provided a method of manufacturing a cordierite porous body using a cordierite forming material including an Al source, an Si source, and an Mg source and forming cordierite by firing, wherein an inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part or all of the Al source and the Si source.

[0010]

In the present invention, a total content of the Si source and the Al source included in the inorganic micro balloon with respect to the whole inorganic micro balloon is preferably 90% by mass or more, when the Si source is converted to SiO_2 , and the Al source is converted to Al_2O_3 . A total content of a sodium compound and a potassium compound included in the inorganic micro balloon with respect to the whole inorganic micro balloon is preferably 2% by mass or less, when the sodium compound is converted to Na_2O , and the potassium compound is converted to K_2O .

[0011]

Moreover, in the present invention, a melting point of the inorganic micro balloon is 1400°C or more, and talc is preferably used as a part or all of the Mg source.

[0012]

[Mode for Carrying Out the Invention]

An embodiment of the present invention will be described hereinafter, but it should be understood that the present invention is not limited to the following embodiment, and modifications, improvements and the like of designs are appropriately added based on usual knowledge of a person skilled in the art without departing from the scope of the present invention.

[0013]

In the present invention, there is provided a method of manufacturing a cordierite porous body using a cordierite forming material including an Al source, an Si source, and an Mg source and forming cordierite by firing, wherein an inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part or all of the Al source and the Si source. Details will be described hereinafter.

[0014]

In the present invention, the inorganic micro balloon for use as a part or all of the Al source and the Si source exhibit a function as a pore former. The inorganic micro balloon has a low specific weight and an appropriate strength as compared with an organic compound based pore former which has heretofore been used, and

therefore is not easily crushed at a mixing/kneading time and easily handled. The inorganic micro balloon exerts a function of a framework which appropriately holds a structure of a formed body before firing, and shrinkage of the formed body during the firing can be suppressed. Furthermore, the inorganic micro balloon is fired to thereby react with the Mg source and the like included in the cordierite forming material, and forms cordierite. That is, since the bubbles of the inorganic micro balloon form a porous structure, a superior pore forming effect is exerted, and the cordierite porous body having a high porosity can be manufactured.

[0015]

Incidentally, since the inorganic micro balloon used in the present invention does not generate any gas component even by firing, it exhibits an effect of inhibiting the resultant cordierite porous body from having defective portions such as cracks, tears and cuts.

[0016]

In the present invention, a total content of the Si and Al sources included in the inorganic micro balloon with respect to the whole inorganic micro balloon is preferably 90% by mass or more, further preferably 95% by mass or more, especially preferably 98% by mass or more, when the Si source is converted to SiO_2 , and the Al source is converted to Al_2O_3 . When the total content of the Si and Al sources is less than 90% by mass, a glass phase is produced, and the balloon is unfavorably easily softened at a lower

temperature. It is to be noted that an upper limit of the content is not especially limited, and theoretically a higher content is preferable.

[0017]

In the present invention, a total content of a sodium compound and a potassium compound included in the inorganic micro balloon with respect to the whole inorganic micro balloon is preferably 2% by mass or less, further preferably 1.5% by mass or less, especially preferably 1% by mass or less, when the sodium compound is converted to Na_2O , and the potassium compound is converted to K_2O . The sodium and potassium compounds contained in the inorganic micro balloon are so-called impurities as viewed from the Si and Al sources (SiO_2 and Al_2O_3) similarly contained in the balloon, and therefore the inorganic micro balloon containing large amounts of these compounds has a low melting point. That is, in the case of use of the inorganic micro balloon (e.g., Shirasu-balloon, etc.) in which the total content of the sodium and potassium compounds contained as the impurities when converted to Na_2O and K_2O , respectively, exceeds 2% by mass, when the firing is performed at around 1420°C , the balloon is quickly and easily molten, the obtained porous body shrinks, and unfavorably the pore forming effect is not exerted well. It is to be noted that a lower limit of the content is not especially limited, and theoretically a lower content is preferable.

[0018]

In the present invention, a melting point of the inorganic micro balloon is 1400°C or more, further preferably 1450°C or more, and especially preferably 1500°C or more. When the melting point of the inorganic micro balloon is less than 1400°C, for example, in the case of the firing at around 1420°C, the balloon is quickly and easily molten, the obtained porous body shrinks, and unfavorably the pore forming effect is not exerted well. It is to be noted that an upper limit value of the above melting point is not particularly limited in the present invention. However, it is preferably 1700°C or less from the viewpoints that the balloon effectively functions as the framework and that the high-porosity porous body is formed. Concrete examples of the inorganic micro balloon which is preferably used in the present invention and which satisfies the above-described various conditions include fly ash balloons (coal ashes) generated as wastes in a thermal power station or the like. It is to be noted that the fly ash balloon is also preferable in that the wastes can be effectively utilized.

[0019]

In the present invention, the average particle diameter of the inorganic micro balloon is preferably 100 μm or less, because a honeycomb having a partition wall thickness of 300 μm or less can be extruded. The average particle diameter is a value measured by a laser scattering type grain size distribution measuring instrument. A compressive strength calculated assuming that the inorganic micro balloon is solid sphere is preferably 1 MPa or more

because the balloon is not easily crushed at a kneading time. The compressive strength is a value measured using the micro compression tester. Furthermore, a tap filling density of the inorganic micro balloon is preferably 0.4 g/cm^3 or less, and a thickness of the shell of the inorganic micro balloon is preferably $10 \text{ }\mu\text{m}$ or less, further preferably $5 \text{ }\mu\text{m}$ or less. It is to be noted that the thickness of the shell is a value measured by observation of a broken or polished face of the shell with a microscope. Concrete examples of the inorganic micro balloon satisfying these conditions include E-SPHERES SL-75 (manufactured by ENVIROSPHERES Co.), but the present invention is not limited to these concrete examples.

[0020]

In the present invention, talc is preferably used as a part or whole of the Mg source included in the cordierite forming material. By the use of talc whose crystal configuration is a plate shape, talc is oriented in the formed body, and, as a result, orientation of a crystal c axis in a direction of extrusion can be imparted to the cordierite after firing, which is preferable because thermal expansion coefficient in a direction of honeycomb extrusion is lowered. It is to be noted that talc for use may contain a trace amount of impurities such as Fe_2O_3 , CaO , Na_2O , and K_2O in a range which does not substantially influence the melting point of the inorganic micro balloon. Compounds other than talc, such as $\text{Mg}(\text{OH})_2$ and MgCO_3 may be used as the Mg source as long as the

effect by the use of talc is not impaired.

[0021]

In the present invention, as to a predetermined component source in the cordierite forming material, there is not especially any limitation except that a specific inorganic micro balloon is used. Details of a method of manufacturing the cordierite porous body of the present invention will be described in accordance with an example of a manufacturing process.

[0022]

First, with respect to 100 parts by mass of the above-described cordierite forming material including the inorganic micro balloon containing SiO_2 and Al_2O_3 at the predetermined ratio, 0.5 to 2 parts by mass of a dispersant, 10 to 40 parts by mass of water, and 3 to 5 parts by mass of a binder as necessary are projected and thereafter kneaded to obtain a plastic clay. Here, the inorganic micro balloon is used as the Al and Si sources, but other materials may be added as the Al source and/or the Si source.

[0023]

Examples of the Al source for use except for the inorganic micro balloon include the source containing one or both of aluminum oxide (Al_2O_3) and aluminum hydroxide ($\text{Al}(\text{OH})_3$) in that impurities are few. Moreover, examples of the binder include hydroxypropyl methyl cellulose, methyl cellulose, hydroxyethyl cellulose, carboxyl methyl cellulose, polyvinyl alcohol and the like. Examples of the dispersant include ethylene glycol, dextrin, fatty

acid soap, polyalcohol and the like. It is to be noted that as to these binder and dispersant, one type alone or a combination of two types or more may be used in accordance with a purpose.

[0024]

Next, the obtained plastic clay is formed into a desired shape, for example, a honeycomb shape or the like by an appropriate forming method. The forming can be performed by an extrusion forming method, an injection forming method, a pressing forming method, a method of forming a ceramic material into a columnar shape and thereafter forming through-holes or the like, and above all the extrusion forming method is preferably performed in that continuous forming is facilitated, cordierite crystals are oriented, and low thermal expansion can be obtained.

[0025]

Next, a raw formed body can be dried by hot air drying, microwave drying, dielectric drying, decompression drying, vacuum drying, or freezing drying, and above all, a drying step of combination of the hot air drying with the microwave drying or dielectric drying is preferably performed in that the whole body can be quickly and uniformly dried.

[0026]

Finally, the dried formed body is preferably fired usually at a temperature of 1410 to 1440°C for three to seven hours depending on the size of the dried formed body. The drying step and the firing step may continuously be performed. The cordierite porous

body can be obtained through the above-described manufacturing process.

[0027]

[Example]

The present invention will be described more concretely hereinafter in accordance with examples, but the present invention is not limited to these examples.

[0028]

(Examples 1 to 4, Comparative Examples 1 to 3)

Inorganic micro balloons indicating values shown in Table 1 with respect to a total content of SiO_2 and Al_2O_3 , a total content of Na_2O and K_2O , and a melting point and talc and Al_2O_3 were fired, and blended at an amount ratio to form a cordierite composition, and a cordierite forming material was obtained. With respect to 100 parts by mass of the material, 2 parts by mass of each of methyl cellulose and hydroxypropoxyl methyl cellulose, 0.5 part by mass of a fatty acid soap as a surfactant, and an appropriate amount of water were added to obtain a clay. The clay was kneaded, and extruded/formed to form a honeycomb structure, and moisture was removed by dielectric drying and hot air drying. Thereafter, firing was performed at a maximum temperature of 1420°C on conditions that a maximum temperature retention time was eight hours, and a cordierite porous body of a honeycomb structure was obtained (Example 1 to 4).

[0029]

It is to be noted that in Table 1, the "total content of SiO_2 and Al_2O_3 " is a value measured by Agglomerated mass plus absorptiometry, and EDTA titration method in conformity to JIS M8853 (fire-resistant clay analysis method), and is a value corresponding to a "total content of Si and Al sources contained in the inorganic micro balloon at a time when the Si source is converted to SiO_2 , and the Al source is converted to Al_2O_3 ". Moreover, a "total content of Na_2O and K_2O " is a value measured by an atomic light absorption process in conformity to JIS M8853 (fire-resistant clay analysis method), and is a value corresponding to a "total content of sodium and potassium compounds included in the inorganic micro balloon at a time when the sodium compound is converted to Na_2O , and the potassium compound is converted to K_2O ".

[0030]

(Example 5)

A cordierite porous body of a honeycomb structure was obtained by a method similar to that of Example 4 except that $\text{Al}(\text{OH})_3$ was used instead of Al_2O_3 (Example 5).

[0031]

(Comparative Example 4)

A cordierite porous body of a honeycomb structure was obtained by a method similar to that of Example 1 described above except that kaolin was used instead of the inorganic micro balloon (Comparative Example 4).

[0032]

(Physical Property Value Evaluation)

The following physical property values were measured with respect to the obtained cordierite porous bodies. The results are shown in Table 1.

[0033]

[Porosity]: measured in Archimedes method

[0034]

[Average pore diameter]: measured by a mercury porosimeter

[0035]

[Coefficient of thermal expansion]: measured by a differential type measuring method using quartz as a standard sample

[0036]

[Permeability]: A sample was prepared by taking a part of a partition wall from each ceramic honeycomb structure to subject to machining in such a manner as to eliminate concave/convex portions. After holding the sample from upside and downside with a sample holder having a diameter of 20 mm in such a manner that there was not any gas leakage, gas was transported under specific gas pressure in such a manner that it is 1 atm on the downstream side of the sample. At this time, as to the gas which passed through the sample, permeability was obtained on the basis of the following equation (1) were measured. In the following equation (1), C denotes permeability (μm^2), F denotes a transported gas flow rate (cm^3/s) measured on the downstream side of the sample, T

denotes a thickness (cm) of the sample, V denotes a gas viscosity (dynes/cm²), D denotes a diameter of the sample (cm), and P denotes a gas pressure (PSI) on the upstream side. In addition, regarding the values shown in the equation, $13.839 \text{ (SI)} = 1 \text{ (atm)}$, and $68947.6 \text{ (dynes/cm}^2\text{)} = 1 \text{ (PSI)}$.

[0037]

(Equation 1)

$$C = \frac{8FTV}{\pi D^2(P^2 - 13.839^2) 68947.6 / 13.839} \times 10^8 \quad \dots (1)$$

[0038]

[Table 1]

	Inorganic micro balloon			Porosity (%)	Average pore diameter (μm)	Coefficient thermal coefficient ^{*2} (×10 ⁻⁶ /°C)	of	Permeability (μm ²)
	Total content (mass%) of SiO ₂ and Al ₂ O ₃	Total content (mass%) of Na ₂ O and K ₂ O	Melting point (°C)					
Example 1	90	1	1500	52	15	1		2
Example 2	95	2	1500	53	14	0.9		3
Example 3	90	3	1400	50	15	1		2
Example 4	98	0.5	1600	66	32	0.8		10
Example 5	98	0.5	1600	67	21	0.3		5
Comp. Ex. 1	89	1	1450	*1	*1	*1		*1
Comp. Ex. 2	95	2.5	1450	*1	*1	*1		*1
Comp. Ex. 3	90	2.5	1350	*1	*1	*1		*1
Comp. Ex. 4	-	-	-	40	8	0.3		0.1

*1: No data (melted upon firing)

*2: Coefficient of thermal coefficient in a direction of honeycomb extrusion

[0039]

As apparent from results shown in Table 1, it has been found that, in the cases (Examples 1 to 4) where an inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part of Al and Si sources included in a cordierite forming material, a cordierite porous body having a sufficiently high porosity can be manufactured in comparison with the cases (Comparative Example 4) where this balloon is not used. It has also been found that the inorganic micro balloon melts upon firing in the cases (Comparative Example 1 to 3) of using an inorganic micro balloon having a total content of SiO_2 and Al_2O_3 of less than 90% by mass and/or having a total content of Na_2O and K_2O of above 2% by mass.

[0040]

It is to be noted that it has been found that either Al_2O_3 (Example 4) or $\text{Al}(\text{OH})_3$ (Example 5) can be used as the Al source other than the inorganic micro balloon.

[0041]

Moreover, it has been found that concerning permeability, as to the cordierite porous body prepared in any of the examples, a value of the permeability is larger than that of the cordierite porous body prepared in Comparative Example 4. Therefore, it is expected that the honeycomb structure prepared in the present embodiment can achieve a low pressure loss and high trapping efficiency when it is used as a material constituting a filter, a catalyst carrier or the like for purifying an automobile exhaust gas.

[0042]

[Effect of the Invention]

As described above, in a method of manufacturing a cordierite porous body of the present invention, since an inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part or all of Al and Si sources included in a cordierite forming material, the method has an advantage that there is a remarkably low possibility of generation of defects such as cuts causing a leakage of a fluid in the obtained cordierite porous body, and a high-porosity cordierite porous body having characteristics preferable for a material constituting a filter, a catalyst carrier or the like mainly for purifying an automobile exhaust gas can be manufactured.

[NAME OF DOCUMENT] Abstract

[ABSTRACT]

[Theme] It is to provide a method of manufacturing a cordierite porous body having a high porosity, in which there is an extremely low possibility of generating defects causing leakage of a fluid, such as cuts.

[Means] A method of manufacturing a cordierite porous body uses a cordierite forming material including an Al source, an Si source, and an Mg source and forming cordierite by firing. An inorganic micro balloon containing SiO_2 and Al_2O_3 is used as a part or all of the Al source and the Si source.

[Adopted Figure] None